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# Slow structural relaxation process facilitates solidification in liquid gallium

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**Slow structural relaxation process facilitates solidification in liquid gallium**F. Demmel <sup>\*</sup>*ISIS Facility, Rutherford Appleton Laboratory, Didcot, OX11 0QX, United Kingdom* (Received 17 October 2019; revised manuscript received 13 January 2020; published 23 January 2020)

The structural relaxation dynamics in liquid and 20 K undercooled gallium has been studied by neutron spectroscopy. The intermediate scattering function at the structure factor maximum was measured over a wide temperature range up to twice the melting temperature. A stretched exponential decay was observed near the melting point which transforms to an exponential decay above  $1.2 T_{\text{melting}}$ . The structural relaxation process beyond 3 ps can be described as a further slow exponential decay. Mode coupling theory is able to calculate the decay rate of this slow structural relaxation, which agrees well with the measurement. This additional structural relaxation, observed in the collective particle motions on next-neighbor distances, can be understood as a slow cage relaxation process, setting in around 370 K and indicating the slowing down of dynamics towards solidification. This process might be the microscopic origin for observed changes in the collective particle dynamics of liquid gallium with decreasing temperature. Liquid aluminium, lead, and rubidium demonstrate a similar behavior, which suggests a universal crossover in dynamics of the equilibrium liquid metal state.

DOI: [10.1103/PhysRevB.101.014207](https://doi.org/10.1103/PhysRevB.101.014207)**I. INTRODUCTION**

Bulk metallic glasses are important materials for technological applications [1,2]. A lot of attention was put towards the understanding of the glass formation process and the ability to form a bulk metallic glass and hence the main research interest was devoted to the materials properties near the glass transition temperature. However, more recently changes in the dynamics above the liquidus temperature in liquid alloys have been reported and moved into the focus of alloy research.

MD simulations on  $\text{CuZr}_2$  demonstrated a violation of the Stokes-Einstein relation at several hundred degrees above the melting point [3]. It was suggested that changes in the collective dynamics are responsible for the variations in the liquid dynamics. Similar observations have been reported in a MD simulation of a  $\text{CuZrAl}$  alloy [4] and *ab initio* based simulations on Al-rich alloys described a breakdown of the Stokes-Einstein relation in the liquid phase [5]. On the experimental side a recent study indicated an Arrhenius crossover in the diffusion coefficients for a wide range of glass forming liquids, including 11 alloys [6]. The crossover temperature was located above the liquidus temperature for the alloys. Also in the binary alloy  $\text{ZrNi}$  a violation of the Stokes-Einstein relation was observed covering several hundred degrees above the melting temperature [7]. All these reports from glass forming alloys demonstrated changes in the dynamics within the liquid state.

For monatomic liquid metals changes with temperature were found in the structural relaxation dynamics on an atomistic length scale for rubidium, lead, and aluminium [8–10]. The amplitude  $S(Q_0, \omega = 0)$  of the collective dynamics at the structure factor maximum  $Q_0$  showed in all three cases a change in the slope in a similar temperature range above the

melting temperature [11]. Furthermore, for all three metals an increase of the generalized longitudinal viscosity was demonstrated which occurs in a temperature range of  $1.3\text{--}1.5 T_m$  upon cooling, suggesting that the liquid metals become more viscous around this temperature range. This temperature range coincides with a slope change of the macroscopic shear viscosity. In liquid rubidium the structural relaxation dynamics transformed from a stretched exponential lineshape to an exponential decay in this temperature range [12,13]. A comparison with mode coupling theory predictions demonstrated that the change in relaxation dynamics is directly connected to a further slow relaxation process, which is believed to be interrelated with structural freezing. A MD simulation with a classical potential on liquid aluminium indicated a breakdown of the Stokes Einstein relation about 200 K above the melting point [14], which agrees with experimental observations [10]. Whether there is a connection to the observed changes in dynamics in the above mentioned glass forming alloys is an open question.

In contrast to the liquid alkali metals, gallium is a low-temperature melting monatomic metal with complex properties. Already the solid phase demonstrates some complexity with a unit cell of eight atoms and pairs of moleculelike dimers [15]. These dimer units might vanish during melting, mirrored in a large melting entropy [16]. Also some pseudopotential implementations show a shoulder, contrasting the simpler potentials of the alkali metals [17,18]. Furthermore, gallium can be undercooled to about  $T \approx 150$  K as a dispersion of small droplets without crystallization [16]. Due to the many curious properties of liquid gallium a large interest in gallium dynamics exists with several studies on the inelastic collective dynamics using neutron and inelastic x-ray scattering [19–23] and also simulations with first-principles methods [24,25]. The ability to undercool gallium attracted early on some interest to study the relaxation dynamic towards solidification with neutron scattering [26,27].

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More recently, we investigated the dynamics of liquid gallium over a wide temperature range from the undercooled state to twice the melting point, focusing on the gallium dynamics above the melting temperature [28]. Changes in the dynamics at next-neighbor distances were observed in a temperature range of about  $1.3\text{--}1.5 T_m$ . Here we present a detailed analysis of the intermediate scattering function  $F(Q, t)$  of liquid gallium at the structure factor maximum over a wide temperature range. The emphasis lies on the evolution of the long lasting part of the structural relaxation process with increasing temperature. A quantitative comparison with predictions from mode coupling theory is achieved.

## II. EXPERIMENTAL DETAILS

The coherent dynamic structure factor  $S(Q, \omega)$  of liquid gallium was measured by neutron spectroscopy [29]. Here we provide only a short description of the experimental and data analysis details. More experimental details can be found in Ref. [28]. Gallium scatters neutrons mainly coherent with a coherent cross section of  $\sigma_{\text{coh}} = 6.675$  barn and an incoherent neutron scattering cross section of  $\sigma_{\text{inc}} = 0.16$  barn [30]. Therefore the neutrons are mostly sensitive to collective particle dynamics, in particular at the structure factor maximum. Quasielastic neutron scattering measurements were performed at the OSIRIS spectrometer of the ISIS Facility, UK. OSIRIS is an indirect time of flight backscattering spectrometer [31]. With a final energy of  $E_f = 7.38$  meV the energy resolution was  $\text{FWHM} = 0.1$  meV. All analysis steps, including empty can subtraction, binning to constant energy and wave-vector bins, have been performed with the Mantid software framework [32]. Finally, the intermediate scattering function  $F(Q, t)$  has been obtained by Fourier transformation and dividing through the Fourier transformed resolution function.

## III. RESULTS AND DISCUSSION

Liquid gallium shows a well-defined deGennes narrowing around the structure factor maximum with a minimum in the linewidth at about  $Q_0 = 2.45 \text{ \AA}^{-1}$  [28]. Figure 1 depicts the intermediate scattering function  $F(Q_0, t)$  of liquid gallium for several temperatures. Included are fits with a stretched exponential decay function. A useful description of liquid dynamics can be obtained by applying the memory function formalism to a generalized Langevin equation [33]. Within the visco-elastic model liquid dynamics is described by a single exponentially decaying memory function [34]. At the structure factor maximum all the relaxation processes are slowed down, well known as deGennes narrowing [35]. At that wave vector the intermediate scattering function  $F(Q, t)$  can be approximated by a single exponential decay function [34]:

$$F(Q, t) = S(Q) \exp(-\gamma(Q)t) \quad (1)$$

with a decay rate of

$$\gamma(Q) = \frac{k_B T Q^2}{m S(Q) M(Q, t=0) \tau(Q)}. \quad (2)$$

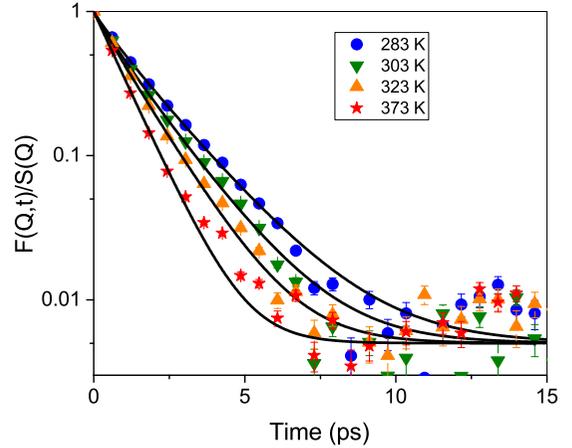


FIG. 1. The Fourier transformed intermediate scattering function  $F(Q_0, t)$  of liquid gallium is plotted on a logarithmic scale for several temperatures against a linear time scale. Included as lines are fits with a stretched exponential function.

Here  $\tau(Q)$  denotes the relaxation time for the exponentially decaying memory function  $M(Q, t)$  within the visco-elastic model. An approximation for the relaxation time, covering the whole wave-number range, was put forward by Lovesey [36]:  $\frac{1}{\tau(Q)} = \frac{2}{\sqrt{\pi}} \{\Omega_L^2(Q) - \Omega_0^2(Q)\}^{1/2}$ . The  $t = 0$  value of the memory function  $M(Q, t = 0)$  is given by  $M(Q, t = 0) = \Omega_L^2(Q) - \Omega_0^2(Q)$ , where  $\Omega_L^2$  denotes the normalized fourth frequency moment of the dynamic structure factor and  $\Omega_0^2 = \frac{k_B T Q^2}{m S(Q)}$  is the normalized second frequency moment [37]. The rate decreases considerably when the main structure factor peak  $S(Q_0)$  is reached, the deGennes narrowing.

Scattering experiments in the energy domain demonstrated that a fit with a single Lorentzian is a good description of the lineshape at the structure factor maximum [28]. However, measurements with good statistics and a wide dynamic range indicated deviations from the single Lorentzian model [12,13]. That the relaxation dynamics beyond the hydrodynamical regime even in simple liquids is more intricate and needs more than one relaxation process was realized in the past. For example, early molecular dynamics simulations on a Lennard-Jones liquid revealed that a memory function based on only one relaxation time is not sufficient to describe the total lineshape properly [38]. Later on, the necessity for at least two times in the memory function was evidenced in modeling the whole dynamic structure factor of several liquid metals [39]. The two time scales in the memory function can be characterized as a fast one, due to uncorrelated binary collisions and a second slower one which is related to collective slow modes. The association with two time scales is made on the memory function level and is not directly linked with the decay of the density correlation function itself.

An often used generalization of the exponential decay function is the Kohlrausch-Williams-Watt (KWW) stretched exponential function:

$$F(Q, t) = A \exp(-t/\tau(Q))^\beta. \quad (3)$$

Here,  $A$  denotes an amplitude,  $\tau$  defines the relaxation time, and  $\beta$  is the stretching parameter. For  $\beta = 1$  one recovers

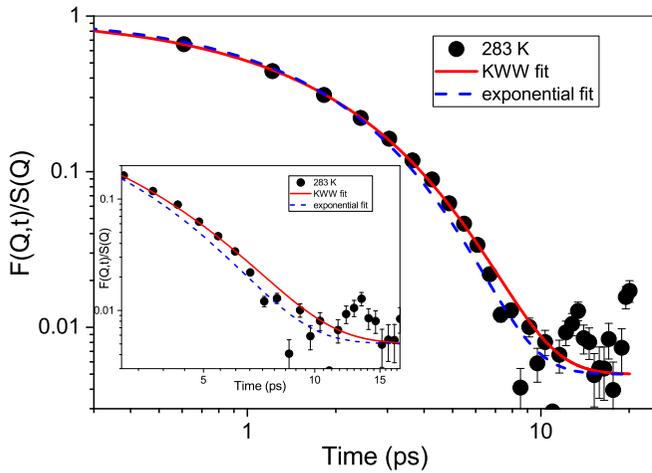


FIG. 2.  $F(Q, t)$  of undercooled gallium at 283 K is presented on a double logarithmic scale. The inset focuses on the small amplitude values. The line depicts the fit with a KWW stretched exponential and the dashed line denotes the fit with an exponential decay.

the exponential decay and a  $\beta < 1$  indicates a stretching and slowing down of the relaxation process.

Figure 2 shows  $F(Q, t)$  of undercooled gallium at 283 K on a double logarithmic scale. Included as a line is the fit with a stretched exponential function and as a dashed line the fit with a simple exponential decay. The inset emphasizes the differences between the two models on the small amplitude level and long times. The smallness of the deviations from the exponential fit confirms the general assumption that a single Lorentzian can describe the lineshape of  $S(Q_0, \omega)$  quite well. Nevertheless, a  $\beta \neq 1$  from the stretched exponential model demonstrates deviations from a simple exponential relaxation. A fit with the KWW function (full line) perfectly describes the whole line shape at  $T = 283$  K with a  $\beta = 0.92 \pm 0.007$  and  $\tau = 1.54 \pm 0.008$  ps. In Fig. 3 we plot the temperature dependence of the line shape parameter  $\beta$ , which shows that  $F(Q, t)$  changes its form with increasing temperature towards an exponential decay at a temperature around 370 K.

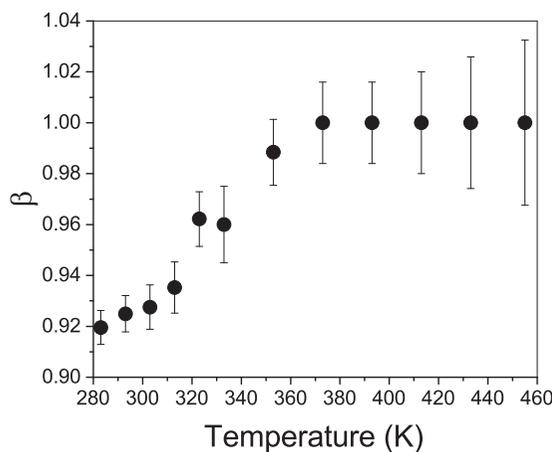


FIG. 3. The  $\beta$  values from the stretched exponential decay fit to  $F(Q, t)$  are plotted against temperature.

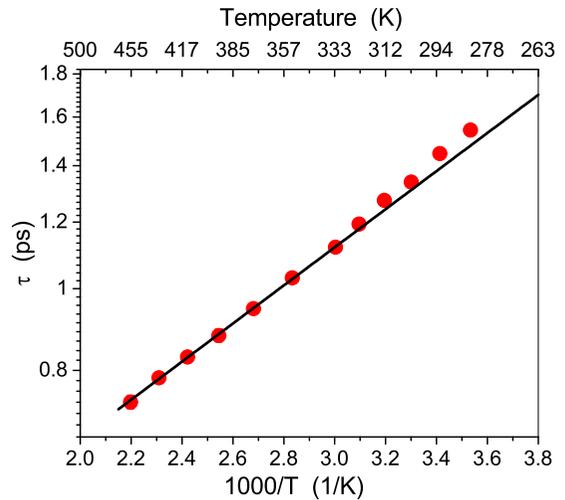


FIG. 4. The relaxation times  $\tau$  are plotted in an Arrhenius-type plot against the inverse temperature. The line is a fit through high temperature points to evidence the changes towards the melting point.

The obtained  $\beta = 0.92$  value for liquid gallium is similar to the one derived for liquid rubidium previously  $\beta = 0.88$  [13] and might indicate that the same structural relaxation process is observed. In metallic alloys, which are known bulk metallic glass formers, stretched relaxation dynamics was also observed. Zr-based alloys demonstrated a stretched exponential function with a  $\beta = 0.8$  [40] and for the Pd-Ni-Cu-P alloy a  $\beta = 0.75$  was reported [41]. In NiNb and NiNbSn alloys  $\beta$  values of up to 0.9 were found [42]. Therefore the stretching in time for gallium is similar to other metals and alloys.

In Fig. 4 the resulting relaxation times  $\tau$  are plotted on a logarithmic scale against the inverse temperature. Through the high temperature points a straight line was fitted to emphasize the changes in the dynamics in this Arrhenius-type plot. Around 340 K the relaxation times deviate to slower times compared to the expected values from the high-temperature extrapolation. The deviation to slower dynamics indicates that a further slow process appears around this temperature. This change in relaxation times accompanies the changes in the lineshape, evidenced through the lineshape parameter  $\beta$ . We conclude from these results and from the previous observations [28] on the amplitude  $S(Q_0, \omega = 0)$  and the generalized longitudinal viscosity  $\eta_l(Q_0)$  that in the temperature range of 1.2–1.5  $T_{\text{melting}}$  the collective dynamics of liquid gallium is altering.

Having demonstrated that the structural relaxation dynamics on a next-neighbor distance changes, we now want to investigate this additional slowing down of the relaxation dynamics in more detail. Mode coupling theory (MCT) is the natural framework to treat such a slow dynamics phenomenon. The Mori-Zwanzig memory function formalism leaves open the definition of the memory function, which is implicitly defined through the fluctuating forces of the surrounding particles. One approach is to use one or more exponential decaying memory functions with relaxation times as parameters. The parameterized memory function with more than one exponential decay function has been applied before, see,

for example, Ref. [39]. However, within MCT the memory function can be related directly to the dynamic structure factor in a self-consistent way. With the assumption that the memory function can be split into a sum of a fast part and a slower contribution the slower part was described by coupling to density fluctuations. Balucani and Vallauri applied MCT to describe the long time decay of  $F(Q, t)$  through an exponential decay [34]. Within some approximations they obtained the following relaxation rate  $\Gamma(Q_0)$ , applicable for the long-time part of the relaxation function [34]:

$$\Gamma = \gamma(Q_0) + 0.5(c + 1)\gamma' - \Gamma_1. \quad (4)$$

Here,  $\gamma(Q_0)$  is the viscoelastic relaxation rate, which is modified by two rates  $\gamma'$  and  $\Gamma_1$ . The decay rate  $\gamma'$  is given by  $\gamma' = \gamma(Q_0)/(1 - \alpha)$ , with  $\alpha = c\gamma(Q_0)\mu$  and  $\mu = (\pi m/k_B T)^{1/2}/2Q_0$ . The parameter  $c = A^2 Q_0 S(Q_0)/8\pi^2 n$  is related to the particle density  $n$  and the wave vector  $Q_0 = 2.5 \text{ \AA}^{-1}$ , where  $S(Q)$  peaks.  $A$  is the area under the main peak of  $S(Q) - 1$ . The second modifying decay rate  $\Gamma_1$  is related to the same parameters in the following way:  $\Gamma_1 = \frac{1}{2}\{(2\gamma + (c - 1)\gamma')^2 + 4c\gamma'^2\}^{1/2}$ .

This solution  $\Gamma$  is valid for long times and neglects the short time evolution, where long times has to be understood as times beyond the decay of the fast visco-elastic relaxation, which is for the case of gallium after about 3 ps. The particle density  $n$  can be calculated from the published mass density data [43]. The area  $A$  under the main peak of  $S(Q) - 1 = 0.52 \text{ \AA}^{-1}$  was obtained through integration of the published 323 K  $S(Q)$  data [44]. For the hard sphere parameter we used  $\sigma = 2.70 \text{ \AA}$ . The first peak in the pair correlation function  $g(r)$  occurs at  $2.8 \text{ \AA}$  [45]. However, such a hard sphere diameter corresponds to a packing fraction of  $\varphi = \pi n \sigma^3/6 = 0.6$ , which is beyond the value when a hard sphere solid melts  $\varphi = 0.545$  [46]. Hence, this  $\sigma$  value can be regarded as an upper limit in a hard sphere description of liquid gallium. For a packing fraction of  $\varphi = 0.545$  we obtain  $\sigma = 2.70 \text{ \AA}$ , which we used as the effective hard sphere diameter of liquid gallium. For the peak value of the structure factor maximum  $S(Q_0) = 2.3$  from the published  $S(Q)$  data is used [44]. The normalized second frequency moment  $\Omega_0^2$  can then be derived.

For the calculation of the normalized fourth moment  $\Omega_L^2$  we applied the approximation given by Hubbard and Beeby [47]:

$$\Omega_L^2(Q) = 3 \frac{k_B T Q^2}{m} + \omega_E^2 \left(1 - 3 \frac{\sin Q\sigma}{Q\sigma} - 6 \frac{\cos(Q\sigma)}{(Q\sigma)^2} + 6 \frac{\sin(Q\sigma)}{(Q\sigma)^3}\right). \quad (5)$$

The important input parameter is here the Einstein frequency  $\omega_E$ . The Einstein frequency describes the average oscillation frequency of a particle surrounded by its neighbors. One way to derive  $\omega_E$  is to calculate the curvature of the potential. However, a wide range of different potential forms exist (see, for example, Refs. [18,48]), which creates a large degree of ambiguity for an estimate of  $\omega_E$ . Therefore, we have chosen a different route to derive an estimate for  $\omega_E$ . The Einstein frequency is directly related to the density of states. For a Debye solid one can show that  $\omega_E = \frac{3}{4} \omega_D$ , because the Einstein frequency represents an average frequency of the vibrational frequencies. From the calculated vibrational

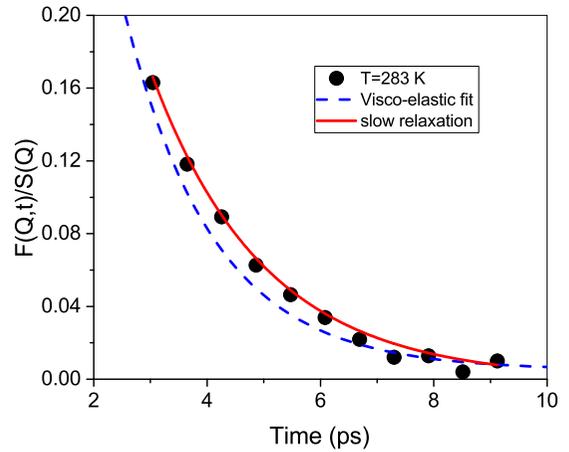


FIG. 5.  $F(Q, t)$  is shown for long times in the relaxation process. The line depicts a fit with an exponential decay function and the dashed line is an exponential fit to the whole relaxation curve, which underestimates the decay of the density fluctuations.

density of states for liquid gallium of an orbital free *ab initio* based simulation [25] we estimate the Einstein frequency as  $\omega_E = 16 \text{ ps}^{-1}$ . That value can be compared to an estimate from the above mentioned recipe with the Debye frequency  $\omega_D$ . The needed Debye frequency for liquid gallium can be approximated by the maximum frequency of the experimentally observed dispersion relation, which is about  $27 \text{ ps}^{-1}$  [23]. Then we arrive at an Einstein frequency of  $\omega_E \approx 21 \text{ ps}^{-1}$ . Therefore we took as an estimate for the averaged Einstein frequency  $\omega_E = 18 \pm 2 \text{ ps}^{-1}$ , where the error indicates the uncertainty in the procedure for the estimate.

With all these parameters the slow relaxation rate  $\Gamma$  can be calculated. For liquid gallium at 283 K we get for the visco-elastic prediction  $\gamma(Q_0) = 0.62 \pm 0.04 \text{ ps}^{-1}$  and the MCT-calculation delivers a relaxation rate of  $\Gamma = 0.48 \pm 0.045 \text{ ps}^{-1}$ .

In Fig. 5 we present  $F(Q, t)$  of gallium in the undercooled state at 283 K for relaxation times longer than 3 ps. At these times the amplitude has already substantially decayed and we are observing values in the 10% range and smaller. The dashed line is a fit to the whole time range between 0 and 20 ps with an exponential relaxation function. That is the expected behavior from the visco-elastic model. The decay rate from the fit delivers  $\gamma(Q_0) = 0.647 \pm 0.006 \text{ ps}^{-1}$  and agrees well with the calculated visco-elastic relaxation rate  $\gamma(Q_0) = 0.62 \pm 0.04 \text{ ps}^{-1}$ . At long times this decay rate does not properly describe the relaxation of the density fluctuations. As a solid line included is a fit with an exponential decay function for times between 3 and 12 ps, which describes the data points quite well. The fit provides a decay rate of  $\Gamma = 0.5 \pm 0.045 \text{ ps}^{-1}$  for the slow relaxation. The fit value and the MCT calculated value agree quite well, taking into account all the approximations.

This slow relaxation process was previously described as the process leading to structural freezing [34]. Our analysis demonstrates that the stretched exponential decay dies away at a temperature around 370 K. Hence, we suggest that this slow relaxation process, eventually leading to solidification, sets in around this temperature upon cooling. This additional

collective motion might be at the origin for the observed changes in the dynamics towards a more viscous behavior of the liquid. The increase of the generalized longitudinal viscosity and the changing slope of the amplitude  $S(Q, \omega = 0)$  might be enabled by this slow structural relaxation process. The amplitude is a measure for the decay of density fluctuations on a next-neighbor distance and the changing slope indicates a more viscous relaxation behavior. We conclude that the collective dynamics in liquid gallium changes at about  $1.2\text{--}1.5 T_m$  from a high temperature more fluid liquid to a more viscous liquid. The range of temperatures is reasoned by the specific quantity under consideration. Amplitude, generalized viscosity, or structural relaxation times are all in different ways more or less directly related to the underlying collective particle movements and hence will evidence the transformation in dynamics in a range of temperatures.

A MD simulation with an embedded atom potential on liquid gallium indicated a change in the dynamics of the self-diffusion coefficient in a similar temperature range [49]. Therefore, in liquid gallium the self-dynamics might also mirror the changes in the collective dynamics. Also the eutectic GaIn alloy demonstrated a crossover of structural and dynamic parameters in a temperature range between 400 K and 550 K [50]. Three monatomic liquid metals, rubidium, lead, and aluminium, showed a very similar evolution of the dynamics with temperature [8–10]. The amplitude  $S(Q_0, \omega = 0)$  and the generalized longitudinal viscosity  $\eta_l(Q_0)$  evidenced a crossover in dynamics in a temperature range of  $1.3\text{--}1.5 T_m$ . MD simulations on liquid rubidium confirmed the changes for the generalized viscosity [51]. Furthermore, a transition from a stretched exponential structural relaxation function to an exponential decay was observed in liquid rubidium [12,13] and liquid lead [9]. The collective inelastic dynamics also provided evidence for a change in dynamics around this temperature range [52]. All these measurements are related to the collective movements of particles and all together draw a picture that the collective dynamics of liquid metals transform to a slower more viscous behavior in a temperature range of about  $1.3\text{--}1.5 T_m$  upon cooling.

The violation of the Stokes-Einstein relation in the experimental study on a ZrNi-alloy [7] was related to a change

in correlated particle movements [53]. MD simulations on a CuZr melt reported a variation in atomic motions far above the melting temperature related to changes in dynamic heterogeneity [3]. A further MD-simulation study on CuZr melts concluded that stringlike collective motions might be at the origin for the observed dynamic heterogeneity and hence for the transformation in dynamics [54]. All these observations emphasize changes in the collective dynamics of the alloys deep in the liquid state. That assessment agrees with the observed transformations in the structural relaxation dynamics for liquid gallium and other monatomic metals. A thermal crossover in the dynamics of dense metallic liquids within the equilibrium liquid state might be a universal behavior.

#### IV. CONCLUSIONS

In summary, an inelastic neutron scattering experiment on liquid and slightly undercooled gallium was performed. The relaxation dynamics at the structure factor maximum was studied up to twice the melting point in small temperature steps. The derived intermediate scattering function  $F(Q, t)$  shows a stretched relaxation behavior near the melting point. With increasing temperature the lineshape transforms to an exponential decay above 370 K. The stretching in dynamics can be understood that an additional slow structural relaxation process sets in. That slow process can be described quantitatively through mode coupling theory and might be the precursor to structural freezing. The temperature range when this transition occurs coincides with a decrease of the generalized longitudinal viscosity. Both facts are evidences that within this temperature range the dense viscous fluid becomes more liquidlike on a microscopic length scale upon heating. The similarity of the liquid gallium behavior with other liquid metals suggests that this transition from a cold-liquid to a hot-liquid response might be a universal feature in liquid metals and possibly also for alloys.

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